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Strong light-matter coupling: parametric interactions in a cavity and free-space

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Abstract. We consider parametric interactions of laser pulses in a coherent macroscopic ensemble of resonant atoms, which are possible in the strong coupling regime of light-matter interaction. The spectrum condensation (lasing at collective vacuum Rabi sidebands) was studied in an active cavity configuration. Parametric interactions under the strong light-matter coupling were proved even in free space. In contrast to bichromatic beats in a cavity, they were shown to appear due to interference between polaritonic wave packets of different group velocities.

Keywords. Quantum optics, dense atomic ensembles, Dicke superradiance, collective effects, resonant parametric processes, polaritons

1. Introduction

We present a study of parametric interactions between laser pulses in a coherent ensemble of two-level quantum objects ("atoms"). The main attention is paid to the processes that can be obtained in the strong coupling regime of light-matter interaction, which is achieved, when (i) the high frequency of field-matter excitation exchanges exceeds the decoherence rates, (ii) the external field is not strong enough to entirely determine the evolution of a system: it is the reemission field that plays a key role and provides the collective behaviour of atoms.

Recently, this regime has attracted attention in quantum optics with both atoms and nanostructures such as quantum wells and dots (cavity QED, Dicke effects, microcavity exciton-polariton parametric scattering, squeezing and entanglement [1]). In both fields, it is considered as one of the key models for quantum information processing (QIP). In this context, the works on single or small number of objects (photons, atoms, excitons) as qubits should be mentioned [2,3].

During last years, there is a growing interest in the study of macroscopic ensembles of quantum objects, which can be used as collective elements for QIP protocols [4,5]. Quantum memories [4,6,7], sources of single photons [8], entanglement of ensembles [9] and new sources of entangled fields based on optically dense atomic ensembles were recently proposed. The advantages of such collective objects in contrast to single-particle qubits were highlighted [10].

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Investigations of the collective effects in the strong coupling regime may be of especial importance, because in this regime, some purely quantum properties of the phenomena do not decrease with increasing number of particles [11,12], which will be important for QIP with both discrete and continuous variables. In Ref. [1], a scheme for generation of twin polaritons (coupled light-matter excitations) in semiconductor microcavities with quantum wells was developed. It was based on the parametric scattering of microcavity exciton-polaritons in the strong coupling regime. In this scheme, very strong quantum correlations between polaritons were obtained inside a cavity. Nevertheless, since only one polariton branch of the dispersion curve was used in the parametric process proposed, quantum correlations in the outgoing light fields were essentially reduced. In this report, we present a study of parametric processes both in a cavity and free space, which arise due to collective energy exchange between field and matter systems (i.e., beating between two polariton branches of the dispersion curve) [13,14,15].

2. Interactions in a cavity

The most significant manifestation of the strong coupling regime consists in the resonant density-dependent splitting of the dispersion curve into two polariton branches. The splitting appears, when the collective coupling constant (cooperative frequency) of a medium ω_c exceeds all rates of incoherent relaxations γ:

$$\omega_c = \sqrt{2\pi d^2 \omega_0 n/\hbar} = g\sqrt{N} \gg \gamma, \tag{1}$$

where d and ω_0 are the dipole moment and frequency of a transition, n and N are the density and number of atoms, g is the single-atom coupling constant. Thus, in dense media, photons with equal wave vectors but different frequencies exist.

If a spatial spectrum of a problem is fixed by a single-mode cavity, beating between these frequencies corresponds to vacuum Rabi oscillations. A spectral doublet, arising in this case, represents density-dependent vacuum Rabi splitting of a cavity mode, which is proportional to ω_c (1). The origin of this effect can be traced to linear interaction of light and dipoles in a passive cavity [12]. The condition of weak field (which cannot destroy polariton dispersion) is reduced to the statement that the photon number is smaller then the number of atoms.

In this report, we present our results related to the strong light-matter coupling in an active system: a multimode broadband laser with an intracavity narrowband coherent resonant medium without population inversion. The condition of the strong coupling (1) can be fulfilled with respect to that macroscopic resonant absorber. A phenomenon of spectrum condensation was analysed. It consists in the dramatic modification of a lasing spectrum under transition from the weak- to strong-coupling regime of light-matter interaction: instead of usual saturated absorption line, a bright narrow doublet of generation appears.

Figure 1 presents dye laser spectra obtained with a neon discharge as an intracavity absorber at different densities n (up to 10^{13} cm^{-3} corresponding to $\omega_c/2\pi$ up to 10 GHz) of neon atoms in the metastable state, which was used as a lower state of a two-level system. It was shown, that the density dependence of the doublet splitting is in agreement with that of ω_c (1), which determines the

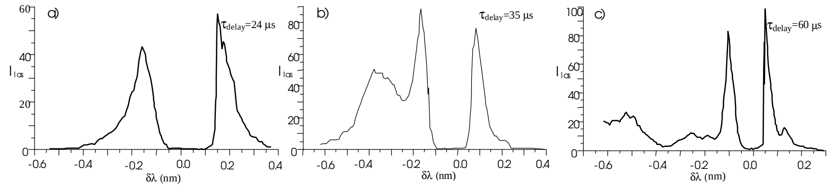

Figure 1. Self-splitting of a dye laser spectrum at different moments of a Ne discharge afterglow, which corresponds to decreasing atomic density in an intracavity cell, $\lambda = 594.5$ nm.

vacuum Rabi splitting. Moreover, the the existence of a density threshold, which is also determined by Eq. (1), was demonstrated. On the basis of the cooperative parametric resonance model [13], this effect was treated as lasing at collective vacuum Rabi sidebands due to parametric interactions between laser modes.

3. Strong coupling regime in free space

The main difference of the free-space interaction consists in the fact that the spatial spectrum of a problem is not fixed by a cavity. As a consequence, under the linear propagation of a short pulse, no coherent density-dependent spectral features (such as vacuum Rabi splitting in a cavity) can be extracted from spectral measurements, except for the trivial appearance of an incoherent absorption line. Nevertheless, in our work, we have shown that such coherent features can be obtained under nonlinear parametric interaction of laser pulses (the details of the theory can be found in Ref. [15], experimental results are presented in Ref. [14]). In contrast to cavity oscillations, they were shown to appear due to free-space optical ringing, which does not originate from beating between waves of equal wave vectors, but from the successive beats between polaritonic wave packets of equal group velocities. Long coherent oscillations can be obtained due to considerable reducing of the group velocity in a dense medium and due to its essential variation over the broad spectrum of a short laser pulse. The oscillation frequency at the initial stage of the ringing ω_D should exceed the decoherence rates γ:

$$\omega_D = \omega_c^2 z/c \gg \gamma. \tag{2}$$

This frequency increases with propagation distance z and medium density n. Thus, it is proportional to the number of interacting atoms, which directly demonstrates the superradiant character of the field reemission by the atomic ensemble.

We considered the nonlinear parametric interaction between two short laser pulses propagating in a dense resonant atomic ensemble. Optical ringing was shown to significantly affect the propagation and amplification of a probe field under its interaction with a nearly copropagating pump. Depending on the probe-pump time delay τ_0 (the probe pulse precedes the pump for $\tau_0 < 0$), the probe transmission spectra show either a specific doublet or dip, which corresponds to parametric sideband amplification or dumping of radiation [cf. Fig. 2(b)]. The widths of these features are greater than the width of an incoherent absorbtion contour [shown in Fig. 2(a)], they are determined by the density-dependent field-matter coupling constant and increase during the propagation [cf. Figs. 2(b), (c)].

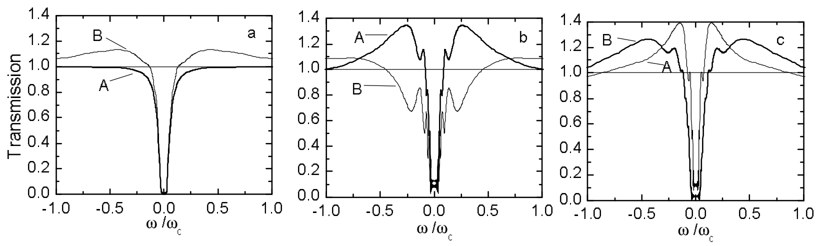

Figure 2. Transmission spectra of the probe field. (a) linear transmission (A) and $\omega_c\tau_0 = 0$ (B), $\omega_c z/c = 1.0$; (b) parametric amplification at $\omega_c\tau_0 = -0.5$ (A) and dumping at $\omega_c\tau_0 = 0.5$ (B) for $\omega_c z/c = 1.0$; (c) parametric amplification at $\omega_c\tau_0 = -0.5$ for $\omega_c z/c = 0.5$ (A) and for $\omega_c z/c = 2.0$ (B); $\gamma/\omega_c = 10^{-3}$, pump area $s_0 = 0.49\pi$.

The spectral features in Fig. 2 can be explained as a result of parametric forward scattering of a probe pulse on spatiotemporal modulations of the population difference, appearing due to the optical ringing in a pump [15]. The characteristic frequencies, which increase with the number of interacting atoms, correspond to the frequency of optical ringing. The condition of the weak fields, which do not destroy dispersion and collective behaviour of atoms, in this case, is equivalent to the requirement of the small input pulse area (so that 0π ringing should not be shadowed by 2π solitons and Rabi flopping). Contrary to strong-field parametric processes (e.g., due to transient Rabi flopping or stationary Mollow-Boyd effect, which are determined by the pump amplitude), the density- and coordinate-dependent spectra of the probe display the importance of free-space collective oscillations and cannot be obtained in the framework of a single-atom model.

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